# High-Pressure Electrical Resistivity Measurements of EuFe<sub>2</sub>As<sub>2</sub> Single Crystals

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#### Abstract.

High-pressure electrical resistivity measurements up to 3.0 GPa have been performed on EuFe<sub>2</sub>As<sub>2</sub> single crystals with residual resistivity ratios RRR=7 and 15. At ambient pressure, a magnetic / structural transition related to FeAs-layers is observed at  $T_0=190\,\mathrm{K}$  and 194 K for samples with RRR=7 and 15, respectively. Application of hydrostatic pressure suppresses  $T_0$ , and then induces similar superconducting behavior in the samples with different RRR values. However, the critical pressure  $\sim 2.7\,\mathrm{GPa}$ , where  $T_0\to 0$ , for the samples with RRR=15 is slightly but distinctly larger than  $\sim 2.5\,\mathrm{GPa}$  for the samples with RRR=7.

## 1. Introduction

Since the discovery of superconductivity in LaFeAs(O,F) with  $T_c = 26 \,\mathrm{K}[1]$ , a family of Fe-pnictide superconductors has attracted much attention. In particular, AFe<sub>2</sub>As<sub>2</sub> (A = Ca, Sr, Ba, Eu, etc.) with a tetragonal ThCr<sub>2</sub>Si<sub>2</sub>-type structure has been intensively studied because of the availability of stoichiometric single crystals with high quality. It turned out that, in Fe-pnictide compounds, the superconducting (SC) ground state could appear in accordance with the suppression of a magnetic/structural transition by doping [2]. In the phase diagrams, it is argued that the superconductivity could coexist and/or compete with the antiferromagnetism [3, 4]. However, a random potential introduced by doping could smear the intrinsic SC properties. For understanding the origin of the high-T<sub>c</sub> superconductivity with T<sub>c</sub> up to 55 K [5], it is of considerable importance to probe the systematic change of ground states using high-quality single crystals. An alternative way to tune the ground state is to apply hydrostatic pressure (P). For instance, recent high-P ac-susceptibility and resistivity measurements have revealed that  $AFe_2As_2$  (A = Sr, Eu) exhibits P-induced bulk superconductivity by suppressing the magnetic/structural transition [6-10]. Meanwhile, superconductivity under hydrostatic Pis absent in CaFe<sub>2</sub>As<sub>2</sub> [11-14], and remains a controversial issue in BaFe<sub>2</sub>As<sub>2</sub> [8, 15-17]

Among the  $A\rm Fe_2As_2$  series, EuFe<sub>2</sub>As<sub>2</sub> is quite unique because the localized Eu<sup>2+</sup> moments order antiferromagnetically at  $T_{\rm N} \sim \! 20\, \rm K$ , in addition to the magnetic/structural transition related to FeAs-layers at  $T_0 \sim \! 190\, \rm K\, [18-21]$ . Interestingly, the magnetic order of Eu<sup>2+</sup> moments

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can be detected even in the SC state induced by doping or application of pressure, which could be a main reason for the novel reentrant-SC-like behavior [9, 10, 22–25].

Here, we report the results of high-P electrical resistivity measurements in EuFe<sub>2</sub>As<sub>2</sub> using newly grown single crystals with a residual resistivity ratio (RRR) as high as 15. At ambient P, the magnetic/structural transition occurs at a higher temperature of  $T_0 = 194$  K, compared with 190 K for single crystals with RRR = 7. Consequently, it is found that the higher quality single crystal requires higher-P to suppress  $T_0$ , and to induce the SC ground state in EuFe<sub>2</sub>As<sub>2</sub>.

## 2. Experimental Details

Single crystals of EuFe<sub>2</sub>As<sub>2</sub> were grown by Bridgman method from a stoichiometric mixture of the constituent elements. In this study, we examined several crystals from two different batches with residual resistivity ratios RRR=7 and 15, where RRR is defined as  $\rho_{300\text{K}}/\rho_{4\text{K}}$ . Single crystals measured in Ref [10] were taken from a batch with RRR=7. High-pressure resistivity measurements of samples with RRR=7 and 15 have been performed simultaneously up to 3.0 GPa using a hybrid-type piston cylinder pressure device [26]. The resistivity was measured by the four-probe method with an ac current I=0.3 mA in the ab-plane. To generate hydrostatic pressure, Daphne 7474 (Idemitsu Kosan) oil, which remains in a liquid state up to 3.7 GPa at room temperature [27], was used as a pressure-transmitting medium. Samples were cooled down in Oxford <sup>4</sup>He system, slowly with an average rate of 0.5 K/min. Applied pressure was estimated at 4.2 K from the resistance change of a calibrated Manganin wire [28].

## 3. Results and Discussions

Figure 1 shows the temperature (T) dependence of electrical resistivity scaled at 300 K ( $\rho/\rho_{300\text{K}}$ ) in EuFe<sub>2</sub>As<sub>2</sub> single crystals with RRR = 7 and 15, where RRR is determined as  $\rho_{300\text{K}}/\rho_{4\text{K}}$ . The measurement was performed in zero field at ambient pressure outside a pressure device with current direction  $I \parallel ab$ . To our knowledge, RRR = 15 is the largest value in EuFe<sub>2</sub>As<sub>2</sub> single crystals [9, 21, 29]. Overall T-variations of the resistivity in the samples with RRR = 7 and 15

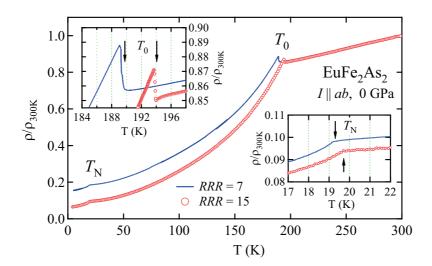
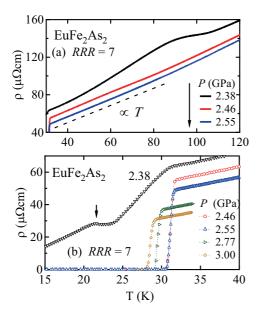
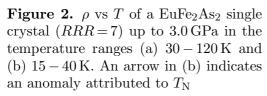


Figure 1. (Color online) The scaled electrical resistivity  $\rho/\rho_{300\text{K}}$  versus temperature in EuFe<sub>2</sub>As<sub>2</sub> single crystals with RRR=7 and 15. The measurement was carried out in zero-field at ambient pressure with the current direction  $I \parallel ab$ . Upper left and lower right insets represent the expanded views around  $T=T_0$  and  $T_N$ , respectively. The data for the sample with RRR=7 in the lower right inset is arbitrarily shifted in vertical direction for clarity.

are qualitatively similar to each other, and are consistent with previous results [9, 20, 21, 29]. It is worthwhile to mention that, as shown in the upper left inset, a magnetic/structural transition temperature  $T_0 = 194 \,\mathrm{K}$  for the sample with RRR = 15 is higher than  $T_0 = 190 \,\mathrm{K}$  for the sample with RRR = 7. This would be the reason why samples with RRR = 15 needs higher pressure (P) to suppress  $T_0$ , as will be discussed below. The Néel temperature  $T_N$  of the localized Eu<sup>2+</sup> moments for the sample with RRR = 15 is slightly higher than the value for the sample with RRR = 7, as can be seen in the lower right inset.

Next, we turn to the pressure effect on the electrical resistivity for the samples with RRR = 7(Fig. 2) and 15 (Fig. 3), which are simultaneously measured in the same pressure device. With increasing P, the resistivity peak related to the magnetic/structural transition is suppressed to a lower temperature in both samples as shown in Figs. 2(a) and 3(a). For the sample with RRR = 7, a reminiscence of the peak is clearly recognized at 2.38 GPa around 100 K, and faintly visible at 2.46 GPa around 70 K as shown in Fig. 2(a). At 2.55 GPa, there is no detectable anomaly, which implies that the critical pressure  $P_c$ , where  $T_0 \to 0$ , may be about 2.5 GPa. For the sample with RRR = 15,  $P_c$  would be  $\sim 2.7$  GPa since the resistivity hump is slightly recognized at 2.69 GPa, but undetectable at 2.77 GPa as shown in Fig. 3(a). It is of interest that the resistivity follows nearly T-linear behavior above  $T_c$  at 2.55 and 2.77 GPa  $(P \sim P_c)$  for samples with RRR = 7 and 15, respectively, as guided by a dashed line. A similar T-variation of resistivity was also reported in several optimally-doped Fe-pnictide superconductors [4, 30–32]. For the sample with RRR = 7, a resistivity upturn and a small maximum, as indicated by an arrow in Figs. 2(b), in the broad SC transition below 31 K are observed at P = 2.38 GPa ( $\langle P_c \rangle$ ). It suggests that the superconductivity is suppressed by the magnetic order of the Eu<sup>2+</sup> moments; consequently, reentrant-SC-like behavior appears. A similar behavior is also slightly seen for the sample with RRR = 15 (Figs. 3(b)), but more smeared out. At  $P > P_c$ , resistivity exhibits sharp SC transitions to zero-resistivity with  $T_c \sim 30 \, \mathrm{K}$  for both samples. With increasing P, the SC





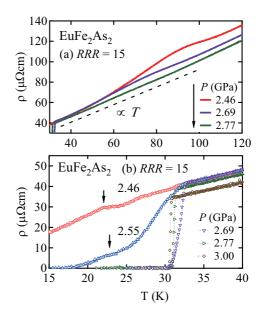


Figure 3.  $\rho$  vs T of a EuFe<sub>2</sub>As<sub>2</sub> single crystal (RRR=15) up to 3.0 GPa in the temperature ranges (a)  $30-120\,\mathrm{K}$  and (b)  $15-40\,\mathrm{K}$ . Arrows in (b) indicate anomalies attributed to  $T_{\mathrm{N}}$ .

transitions persist up to 3.00 GPa although the  $T_{\rm c}$  continuously decreases. Thus, the P-variation of the resistive behavior between the samples with different quality is qualitatively similar to each other, and is consistent with the previous result [10]. However,  $P_{\rm c} \sim 2.7$  GPa for the sample with RRR=15 is slightly but distinctly larger than  $\sim 2.5$  GPa for the sample with RRR=7, which may be as a consequence of the larger value of  $T_0$  for the higher-quality sample at ambient-P. We have repeated similar high-P resistivity measurements using several single crystals, and confirmed that the observed difference in the magnitude of  $T_0$  and  $P_c$  between the samples with RRR=7 and 15 is beyond the error of the pressure estimation ( $\pm 2$ -3×10<sup>-2</sup> GPa) [28]. Another meaningful issue, which probably relates to the sample quality, is the width of a SC transition  $\Delta T_c$ . The minimum values of  $\Delta T_c$  are 1 K and 0.8 K for samples with RRR=7 and 15, respectively. These facts suggest that the higher-quality single crystals have larger values of  $T_0$  and  $P_c$  as well as a sharper SC transition in EuFe<sub>2</sub>As<sub>2</sub>.

Until now, there has been no report concerning the quantum oscillation in EuFe<sub>2</sub>As<sub>2</sub>, despite the importance for understanding the Fermi surface topology and mass renormalization. In fact, we have already tried de Haas-van Alphen (dHvA) measurements of EuFe<sub>2</sub>As<sub>2</sub> using the samples with RRR = 7 at 0.6 K with fields up to 35 T, but could not detect any dHvA oscillation. Given that quantum oscillations were successfully detected in SrFe<sub>2</sub>As<sub>2</sub> ( $RRR \sim 8$ ) [33] and BaFe<sub>2</sub>As<sub>2</sub>(RRR = 10) [34], it is worthwhile to perform the dHvA measurement of EuFe<sub>2</sub>As<sub>2</sub> using the newly grown single crystals with RRR = 15.

#### 4. Conclusions

We have performed high-pressure electrical resistivity measurements up to 3.0 GPa in EuFe<sub>2</sub>As<sub>2</sub> single crystals with RRR=7 and 15. At ambient pressure, a magnetic/structural transition occurred at  $T_0=190\,\mathrm{K}$  and 194 K for the samples with RRR=7 and 15, respectively. Although P-induced superconductivity was confirmed in the samples with different RRR values, the critical pressure  $P_{\rm c}\sim2.7\,\mathrm{GPa}$  for the samples with RRR=15 was slightly but distinctly larger than  $\sim2.5\,\mathrm{GPa}$  for the samples with RRR=7.

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